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Electrodialytic Remediation of Soil Fines (< 63µm) in Suspension - Influence of Current Strength and L/S.

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ABSTRACT

Current treatment of the remaining soil fines from soil wash is onerous and expensive, and therefore, in this work, we investigated the feasibility of electrodialytic remediation (EDR) as an alternative treatment. The study focuses on EDR efficiency as a function of current strength, liquid-to-solid-ratio (L/S), pH and time. We found out that during the experiments, Pb was easily dissolved by the acidification resulting from water splitting at the anion-exchange membrane. When higher currents and/or higher L/S ratios were applied, it was found that water splitting occurring at the cation-exchange membrane increased the pH, and this resulted in decreased remediation efficiency. It was shown that complete remediation of the soil-fines is possible, with the majority of the Pb being transported into the catholyte and precipitated at the cathode. Based on the results it is recommended that EDR is implemented using a number of reactors in series,

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where the initial reactor works at the highest possible removal rate, and the final reactor works at the target Pb-concentration.

KEYWORDS: Electrodialysis; Pb; soil remediation; soil washing; water splitting.

1 INTRODUCTION

1.1 Soil washing

Particle separation techniques based on size or density differences are standard operations in the supply of clean sand for concrete, road-building and in mining technology . Variations of such techniques namely soil washing have been investigated for their potential application in remediation of contaminated soil , and constitute one of few options in the treatment of heavy metal (HM) contaminated soil. Through soil washing, the oversize material can be cleaned simply by water-rinse. The fine and coarse sands can be treated by density/gravity separation processes, followed by an extractive soil washing where an appropriate extractant is added. The remaining silts and clays, which generally contain the highest concentrations of contaminants, are dewatered and treated by stabilization/solidification techniques to immobilize the contaminants . However, fines are dewatered with difficulty: minimum 45% water-content after thickening and pressurized belt filter press . Including a volume increase due to the stabilization/solidification-process, the final volume of contaminated material may well resemble the initial even though a considerable volume of clean materials has been obtained. The limited success of soil washing can largely be attributed to this troublesome treatability and handling of the fine fraction. Attempts have been made

to use the contaminated fines for brick- and roof-tile fabrication for which clay is a natural raw material. However, legacy- and confidence matters have restrained this solution . An introduction of an efficient unit-process for decontamination of the fines is necessary to make soil washing an environmentally and economically profitable process, where contaminated soil is remediated and well-defined materials for construction-purposes are produced.

With the objective to develop a method for treatment of the remaining fines from soil washing, this work aims at investigating the feasibility of Electrodialytic remediation (EDR) of Pb-contaminated fines in suspension. Influence of L/S and current were identified as important, basic parameters, and therefore this work focuses on elucidation of remediation dependency on these.

1.2 EDR of Pb from fine-grained material in suspension

EDR is an electrokinetic remediation method, where ion-exchange membranes function as barriers, which physically hinder intrusion of hydrogen and hydroxide-ions from the electrode processes into the contaminated material. EDR of HM contaminated soils has up till now been tested only for stationary set-ups. However, EDR of fine materials was already tested in suspension (non-stationary set-up) for fine-grained material, such as municipal solid waste incineration (MSWI) fly ash , wood combustion fly-ash , straw combustion fly-ash , wastewater sludge and contaminated harbor sediments . These materials are difficult to handle in solid form, and therefore the suspended EDR setup was introduced .

In MSWI fly ash, Pb was found to be the least mobile of the contaminating metals, and only 8% was removed after 3 weeks of remediation (0.796 mA/cm^2 and

L/S-ratio 6.5) . The low removal led to introduction of desorbing agents, such as sodium-citrate (12% removal after 2 weeks for the same ash and experimental conditions) and ammonia/ammonium citrate (20% removal after 70 days) . From a different fly ash, 30.7% Pb was removed after 12 days (0.8mA/cm^2 and L/S 5.25) . From contaminated harbor sediments, Pb was removed more efficiently: 76% Pb was removed after 21 days (0.6 mA/cm^2 and L/S 4) in distilled water . Remediation was shown to depend on L/S-ratio and current-strength as well as time, and 91-96% was removed from various harbor sediments after 14 days (1.393mA/cm^2 and L/S 8) .

1.3 Ion-exchange membranes and water splitting

For all the fine-grained materials referred above, as well as in EDR of soil in stationary set-up , water splitting was observed at the surface of the anion-exchange membrane. The occurrence of water splitting is well known from dialysis of liquid samples , and *accelerated water splitting* is used for electro-synthesis of acids and bases in industry. *Accelerated water splitting* is obtained through application of bipolar ion-exchange membranes at the surface of which the dissociation rate is 10^7-10^8 times faster than in free solution . The presence of soil also affects the water splitting rate positively e.g. application of iron hydroxide/oxide and silica sol at cation-exchange membranes increased the rate up to 10^4-10^5 times . Due to the negatively charged surface of soil-constituents such as clay and organic matter, the interface between an anion-exchange membrane and soil constituents is in effect bipolar, and very little cation-exchange

capacity or clay is necessary for water splitting to occur at the bipolar interface between anion-exchange membrane and soil .

Initiation of water splitting is related to the limiting current (i_{lim}). As current is increased, the ion-concentration at the surface of the membrane approaches zero (concentration polarization), and i_{lim} is reached:

$$i_{lim} = \frac{zDFc_b}{\delta (t^m - t^{bl})} \quad (1)$$

Here z is the charge, F is Faraday's constant, c_b is the ion-concentration in the bulk solution, δ is the boundary layer thickness. t^m and t^{bl} are the transport numbers of the counter-ions transported in the membrane and the boundary layer respectively, and D is the diffusion coefficient of the counter ions in the boundary layer . A region exists, where voltage is increased dramatically with an increase in the current. In this region the additional energy (ΔV) is a result of increasing resistance associated with the concentration polarization in the boundary layer at the polarized membrane side . The limiting current density increases with increasing ionic concentration (c_b) and decreases with thickness of the boundary layer (δ). Consequently the limiting current increases with decreased L/S-ratio (more ions available). Furthermore i_{lim} increases in a stirred system compared to a stationary set-up because the boundary layer is decreased. In EDR of soil in stationary systems, the limiting current density for the cation-exchange membrane was in two cases found to be around 0.4 mA/cm^2 , while the limiting current for the anion-exchange membrane is close to zero. The water splitting at the anion-

exchange membrane results in acidification of the soil with dissolution of HM's as a consequence. This acidification is the foundation of unenhanced EDR of HM-containing materials . Water splitting at the cation-exchange membrane is in most cases unwanted due to production of hydroxide-ions and decreased mobility of most HM's under alkaline conditions, why the ideal current density is found just below $i_{\lim}$ for the cation-exchange membrane.

2 MATERIALS AND METHODS

2.1 Soil

The experimental soil is an industrially contaminated Danish soil of unknown origin, characterized in as soil 10 to be rich in carbonate and feldspar, low in organic matter and relatively rich in phosphate. SEM-EDX analysis revealed a mixed Pb-pool, where Pb was identified in association with iron/aluminum-minerals, metallic alloys, solder, chloride and pure (possible metallic) Pb .

The soil-fines were obtained by simple wet-sieving of the original soil with distilled water through a 0.063mm sieve. A concentrated slurry of fines was obtained by centrifugation at 3000rpm for 10 min. followed by decantation of the supernatant. The soil-fines were kept as slurry and stored at 5ºC in presence of oxygen. Prior to an experiment, distilled water was added to the slurry to obtain the wished L/S-ratio.

2.2 Characterization

The original soil as well as the fines were analyzed for the following parameters: **Metals** (Fe, Pb) were measured by AAS/graphite furnace. Soil samples were digested according to the Danish standard method DS259 , which includes acid digestion of 1g soil with 20.00mL of half concentrated HNO_3 in autoclave at 200kPa and 120ºC for 30 minutes and filtration through a 0.45µm filter. Liquid samples with pH > 4 were digested with one part of conc. HNO_3 to four parts of liquid in autoclave at 200 kPa and 120ºC for 30 minutes prior to AAS measurement. **Carbonate** content was determined volumetrically by the Scheibler-method when reacting 3g of soil with 20mL of 10% HCl. The amount was calculated assuming that all carbonate is present as calcium-carbonate. **Organic matter** was determined by loss of ignition in a heating furnace at 550ºC for 1 hour. **CEC** was decided by ion exchange of 10g dry soil with NH_4^+, followed by exchange of NH_4^+ for Na^+. The ammonium concentration of the centrifugate was measured by spectrophotometer via flow-injection. **Phosphate** was measured after digestion of 0.2-0.5g sample at 550ºC followed by boiling with HCl. The sample was reacted with ammonium molybdate to form yellow phosphor-molybden acid, which was reduced by ascorbic acid in the presence of antimony. The strong blue color was measured by spectrophotometer Shimadzu UV-1601. **pH** was measured by electrode MeterLab® CDM220 after shaking of 5.0g dry soil with 12.5mL 1M KCl constantly for 1 hour, followed by settling for 10min. **Pb concentration in each grain-size fraction** was measured according to DS 259 after wet-sieving (with 0.002M $Na_4P_2O_7$) approximately 100g naturally wet soil through a 0.063mm sieve followed by separation by dry sieving of the larger fractions (>0.063 mm). Each fraction was grounded thoroughly with a mortar and pestle in order to obtain a

homogeneous distribution of Pb. **Desorption dependency on pH**: 5.00g of dry, crushed soil was mixed with 25.00ml reagent at a shaking table at 200rpm for 7 days. pH was measured after 10min settling, and the liquid was filtered through a 0.45µm filter for subsequent measurement on AAS. The reagents were: 1.0M NaOH, 0.5M NaOH, 0.1M NaOH, 0.05M NaOH, 0.01M NaOH, distilled water, 0.01M HNO_3, 0.05M HNO_3, 0.1M HNO_3, 0.5M HNO_3, 1.0M HNO_3. **Sequential extraction** was made according to the method from the Standards, Measurements and Testing Program of the European Union : 0.5g of dry, crushed soil was treated in four steps as follows: I) Extraction with 20.0ml 0.11M acetic acid pH 3 for 16 hours. II) Extraction with 20.0 ml 0.1M $NH_2OH{\cdot}HCl$ pH2 for 16 hours. III) Extraction with 5.0ml 8.8M H_2O_2 for one hour and heating to 85ºC for one hour with lid followed by evaporation of the liquid at phase 85ºC until it had reduced to < 1ml by removal of the lid. The addition of 5.0 ml 8.8M H_2O_2 was repeated followed by resumed heating to 85ºC for one hour and removal of the lid for evaporation until almost dryness. After cooling down, 25.0 ml 1M NH_4OOCCH_3 pH 2 was added, and extraction took place for 16 hours. IV) Finally digestion according to DS 259 was made for identification of the residual fraction. Between each step the sample was centrifuged at 3000rpm for 15min, and the supernatant was decanted and stored for AAS analysis. Before addition of the new reagent the sample was washed with 10.0ml distilled water for 15min, centrifuged at 3000rpm for 15min and the supernatant was decanted. All extractions were performed at room temperature while shaking at 100rpm unless otherwise mentioned. All analyses were made in triplicate except CEC and sequential extraction which were made in double.

2.3 Remediation experiments

Electrodialysis experiments were made in cylindrical Plexiglas-cells with three compartments. Compartment II, which contained the soil-slurry was 10 cm long and 8 cm as an inner diameter. The slurry was kept in suspension by constant stirring with plastic-flaps attached to a glass-stick and connected to an overhead stirrer (RW11 basic from IKA). The anolyte was separated from the soil specimen by an anion-exchange membrane, and the catholyte was separated from the soil specimen by a cation-exchange membrane. Figure 2.1 shows a schematic drawing of the setup. Both membranes were obtained from Ionics® (types AR204SZRA and CR67 HVY HMR427). Electrolytes were circulated by mechanical pumps (Totton Pumps Class E BS5000 Pt 11) between electrolyte chambers and glass bottles. Platinum coated electrodes from Permascand® were used as working electrodes, and the power supply was a Hewlett Packard® E3612A. The electrolytes initially consisted of each 500mL 0.01 M $NaNO_3$ adjusted to pH 2 with HNO_3.

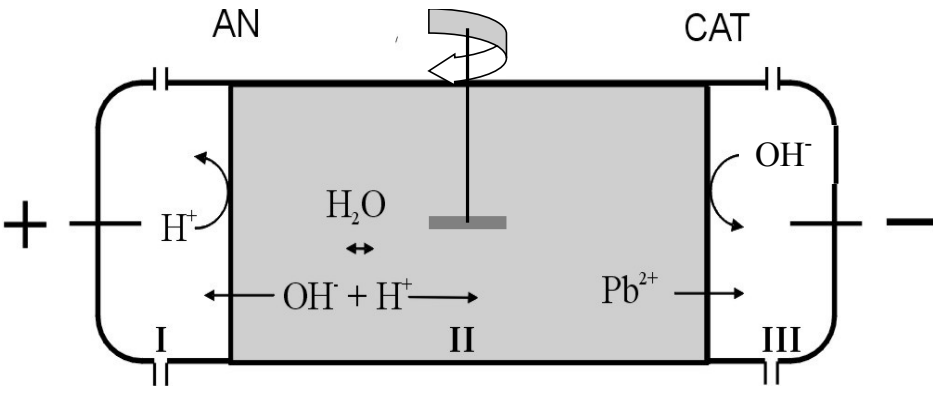

Figure 2.1: Schematic view of a cell used for experimental EDR remediation of soil-fines in suspension. AN = anion-exchange membrane. CAT = cation-exchange membrane.

Current, voltage and pH in all chambers as well as conductivity in chamber II, were measured approximately once every 24 hours. During the electrodialysis experiments current passed between the electrodes. Due to electrode processes pH-changes occurred in the electrolytes, and pH in the electrolytes was manually kept between 1 and 2 by addition of HNO_3 and NaOH.

To investigate the influence of current strength and L/S, 12 experiments were made according to table I. It was earlier shown that Pb in this soil and most other soils desorb at pH < 2, why all experiments were run until pH in chamber II was decreased to < 2 by the water splitting process.

TABLE I

Experimental plan

L/S Current density (J) [mA/cm^2 membrane area]	3.5	7.0	10.5	14.0
0.2	A1	A2	A3	A4
0.4	B1	B2	B3	B4
0.6	C1	C2	C3	C4

Apart from these 12 experiments, 2 additional experiments were made (C1a and D1a). These experiments worked at L/S 3.5, while running for the same amount of time pr. g. soil as experiment C2. Constants and variables of the four compared experiments are summarized in table II.

TABLE II

Constants and variables of two additional experiments and the experiments with which they are compared.

Experiment	C1	C1a	C2	D1a
Current density (*i*) [mA/cm^2 membrane area]	0.6	0.6	0.6	0.8
L/S	3.5	3.5	7.0	3.5
Treatment-time (hours/gram soil)	6.0	11.5	11.5	11.5

3 RESULTS AND DISCUSSION

3.1 Soil characteristics

Characteristics of the original soil and the soil fines are listed in table III. The soil fines contain more Pb, carbonate, organic matter and iron than the original soil. Also CEC is higher, probably due to higher fraction of organic matter and clay-minerals. In contrast the phosphate-content is lower.

TABLE III

Characteristics of the soil fines and the original soil

	Pb [mg/kg]	**pH**	**$CaCO_3$ [%]**	**Organic matter [%]**	**CEC [meq/100g]**	**PO_4^{3-} [mg/kg]**	**Fe [g/kg]**
Soil fines	1170	7.8	17.3	7.8	14.1	559	27.3
Original soil	1090	7.8	9.2	2.8	4.5	1547	13.6

Figure 3.1 reveals that Pb is concentrated in the < 63µm fraction as well as in the fraction between 0.25 and 1mm. In this larger fraction Pb is unevenly distributed (large standard deviation on analysis of Pb), and may well be found as discrete particles of contaminating metal, which may be separated from the soil matrix by density-separation during soil washing. It should be stressed that no attempts have been made to optimize the soil washing process, and an even more pronounced concentration in the fines could be expected if this was done.

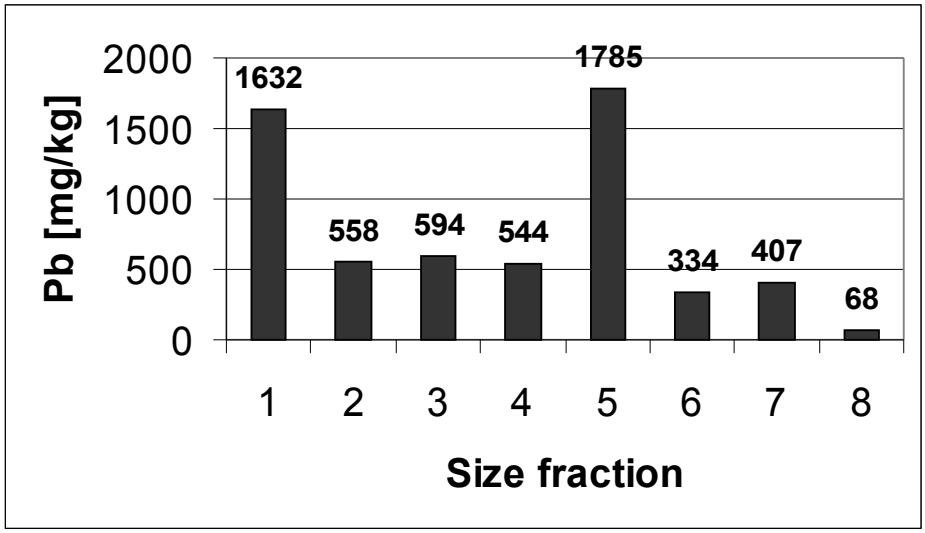

Figure 3.1: Distribution of Pb in soil fractions. 1: 0-0.063mm; 2: 0.063-0.080mm; 3: 0.080-0.125mm; 4: 0.125-0.250mm; 5: 0.250-1.000mm; 6:1.000-2.000mm; 7: 2.000-4.000mm; 8: > 4.000mm.

In figure 3.2, the desorption dependency on pH from the original soil and the soil fines is illustrated. At pH below 2 most of the Pb is desorbed from both materials, and their extraction-patterns are very similar.

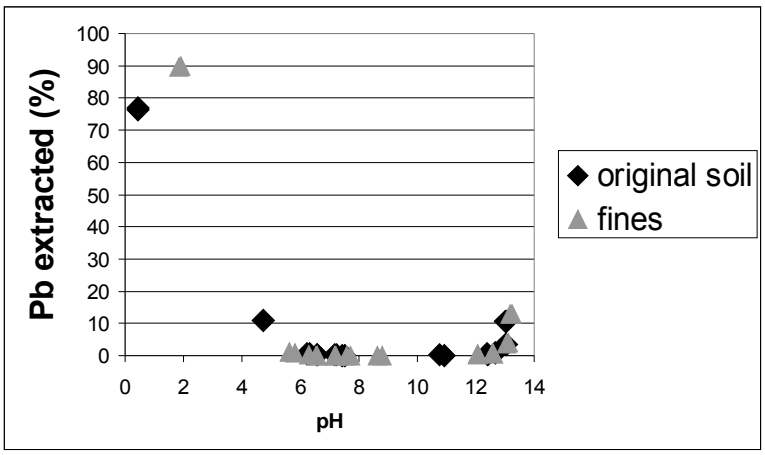

Figure 3.2: Desorption dependency of Pb in original soil and soil fines.

Sequential extractions of the soil-fines and original soil show (figure 3.3) a large residual fraction of Pb in the original soil, while most of the Pb in the fines is released during oxidization. The higher mobility of Pb in the fine fraction suggests that Pb in the coarse fractions may exist in stabile e.g. metallic compounds. As the stabile compounds are slowly transformed under environmental conditions, the Pb may preferably bind to organic matter and clay-particles prevailing in the fine fraction.

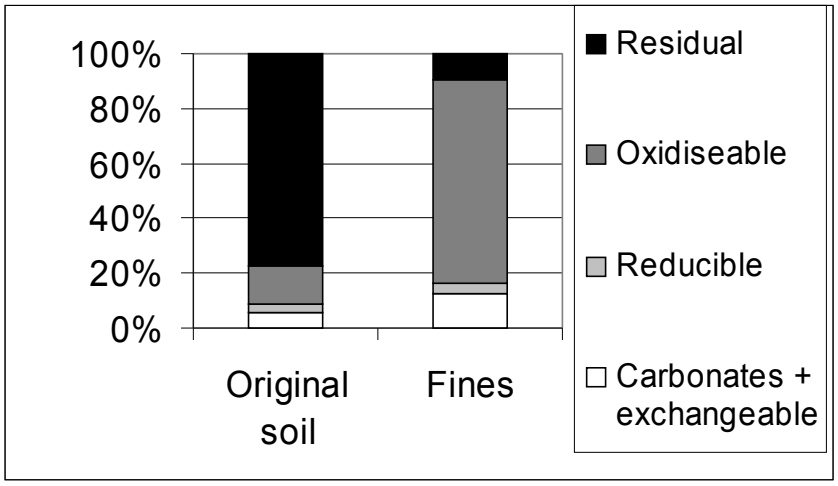

Figure 3.3: Sequential extraction of Pb from soil fines and original soil.

3.2 EDR experiments

Table IV gives results of the 12 experiments listed in table I.

TABLE IV

Results of EDR experiments.

Best results in each series are emphasized by bolding.

Exp. No.	Mass-balance [%]	Hours to reach pH < 2	Hours pr. g soil	Current efficiency [‰]	Pb removed from II [%]	Pb in liquid in II [%]	Hours pr. mg Pb removed	Final Pb soil conc. [mg/kg]
A1	126	713	**6.9**	0.4	21	43	29.5	681
A2	130	403	7.5	0.5	25	35	22.9	753
A3	125	330	8.9	1.0	63	14	9.9	407
A4	134	265	9.5	**1.2**	72	5	8.5	426
B1	113	495	**4.8**	0.4	32	38	13.9	584
B2	133	334	**6.2**	0.6	51	21	8.2	506
B3	115	240	**6.5**	**0.9**	**86**	**0.6**	**5.6**	**224**
B4	161	474	17.0	0.5	**87**	**1.6**	10.4	**212**
C1	159	621	**6.0**	**0.7**	72	15	**4.6**	309
C2	87	621	11.5	0.3	**90**	**0.7**	12.7	**119**
C3	132	788	21.3	0.2	**96**	**1.4**	14.3	**42**
C4*	91	809	28.9	0.1	**93**	**0.0**	29.3	**133**

*Reached maximum voltage (137.1V) after 330 hours and continued with variable current density (0.3-0.5mA/cm^2) until 447hours, where the current density increased to 0.6mA/cm^2 again and remained stabile. pH in this experiment did not reach 2 when the experiment was terminated.

3.2.1 Mass balances

Mass-balances for Pb were obtained between 87 and 161%. Since the initial Pb-concentration was determined as a triplicate measurement in a batch of soil-slurry, containing slurry for several experiments, and the final amount was specifically calculated for the single experiment, the final concentration is regarded the most precise, and therefore used when calculating removal percentages. In each experiment a weight-loss approximately equivalent to the amount of $CaCO_3$ in the soil was observed.

3.2.2 Water splitting and current efficiency

In the A-series of experiments, the time to reach pH <2 decreased with increased L/S, as expected, due to presence of less soil and therefore less buffer capacity of the slurry. In the B-series and even more pronounced in the C-series however, other mechanisms influence the acidification of the slurry. In the B-series time to reach pH <2 was decreasing from experiment 1 through 3; while increasing dramatically in experiment 4. In the C-series time to reach pH <2 is increased compared to the B-series although current was increased, and within the series itself an increase was observed throughout the whole series. The effect was even more pronounced when calculated as hours to reach pH <2 pr. g of soil. The reason for this increase we believe is the exceeding of the limiting current-density for the cation-exchange membrane resulting in production of hydroxide-ions. The lack of ions becomes more pronounced as L/S and current increases due to less soil material to supply the ions and more ions necessary for transport. With production of both hydrogen and hydroxide-ions in the cell, acidification was impeded. In addition, part of the current was transported by hydrogen and

hydroxide-ions, resulting in a decreased current efficiency (‰ of the charge carried by Pb^{2+}) as seen for experiments B4 and C2-C4. The described processes became more pronounced the more current was forced at the system. In general however, current efficiency was approximately 10 times larger than when remediating harbor sludge probably because the soil contains less soluble salts competing for the current. Experiment C4 is an example of the utmost consequence of forcing too much current over the system: the lack of ions became pronounced, and even water splitting was unable to compensate. As a result resistance increased dramatically, and the constant current-density could not be kept.

The mechanisms are illustrated in detail in figure 3.4 showing the conductivity of the soil-slurry as a function of time; figure 3.5 illustrating the pH-development and figure 3.6 showing the voltage-development. After a “lag-period”, conductivity increased in all experiments except C4. The extent of the “lag-period” was related to the current-density and L/S relationship with a longer “lag-period” for experiments with high current density and high L/S. The conductivity of the soil-slurry in all experiments in the A-series, as well as B1-B3 and C1 started to increase after approximately 150 hours. While the conductivity increase for experiment B4, C2 and C3 appeared after almost 400, 450 and 620 hours respectively and the conductivity increase in C4 was never observed.

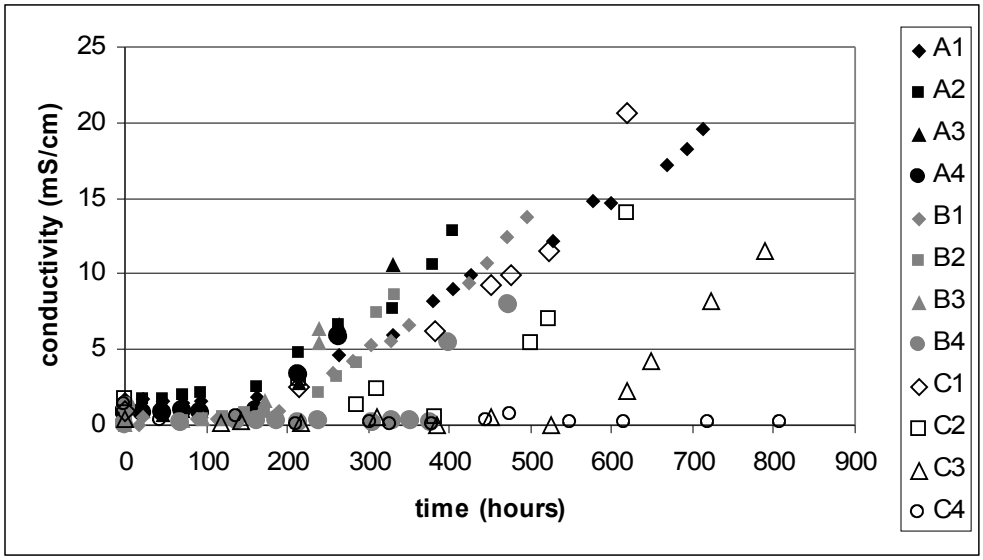

Figure 3.4: Conductivity in the soil-slurry of the 12 experiments as a function of time.

In figure 3.5 the pH-development is illustrated. All experiments except C4 reached <2, and were then terminated. As already discussed, the time to reach pH<2 within each series of experiments depends on the L/S-ration for experiments not affected by water splitting at the cation-exchange membrane (A1, A2, A3, A4, B1, B2, B3). During the lag-phase we believe that soluble salts including carbonates were removed from the soil. As the buffer-capacity was spend, the excess production of H^+-ions in the soil-slurry resulted in the simultaneous pH-decrease and conductivity increase.

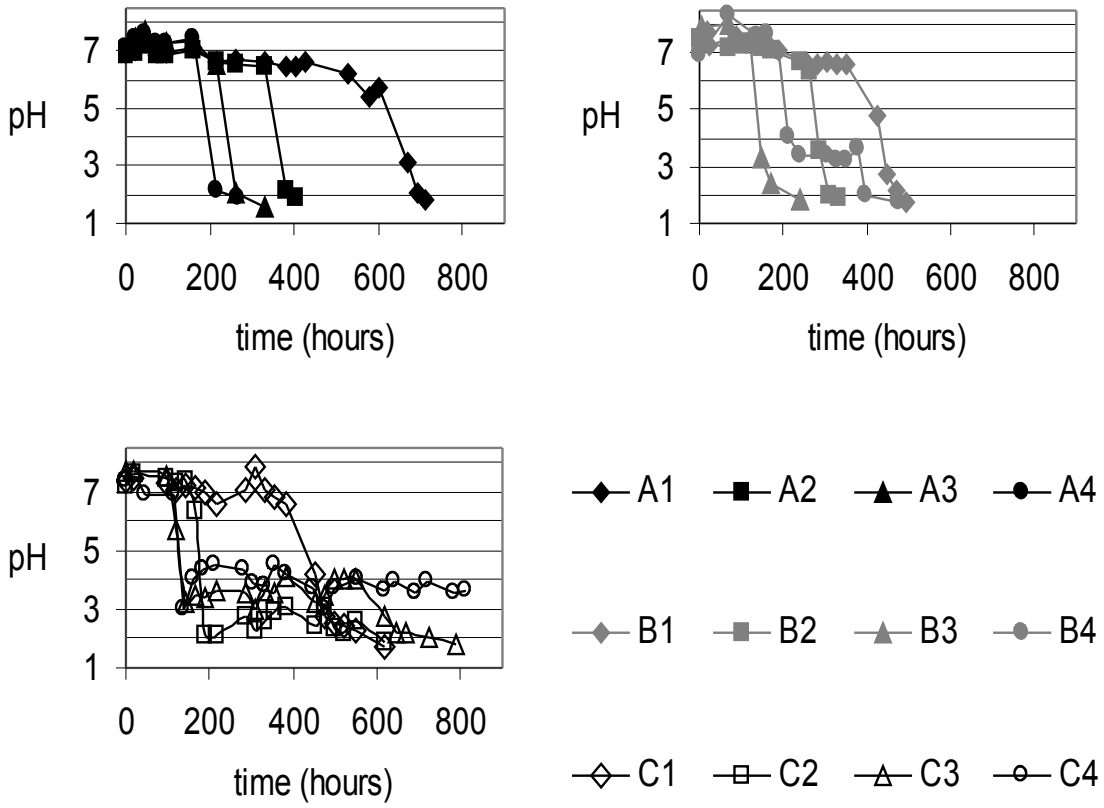

Figure 3.5: pH-development in the 12 experiments as a function of time.

In figure 3.6 it is illustrated how the voltage in all experiments in the A-series was low (<10V) throughout the experimental time. Experiment B2, B3, C1 and C2 showed a few incidents of high voltage caused by precipitation of hydroxides in the cation-exchange membrane in connection with high pH in the catholyte. In these cases voltage decreased immediately after pH-adjustment. In comparison the voltage-increases in experiments B4, C3 and C4 were more constant and not related to pH-increases in the catholyte. These voltage increases we believe appear due to extensive concentration polarization and water-splitting at the anion-exchange membrane, and the observations suggest that under continuous pH-

control, voltage can be used as a control-parameter for avoidance of water splitting at the cation-exchange membrane. In this experimental setup water splitting could be avoided by keeping a voltage-drop between the working electrodes below approximately 20. It should however be stressed that the major voltage-drop in that case is found over the membranes and that reactor up-scaling therefore cannot be made with a simple linear voltage-increase.

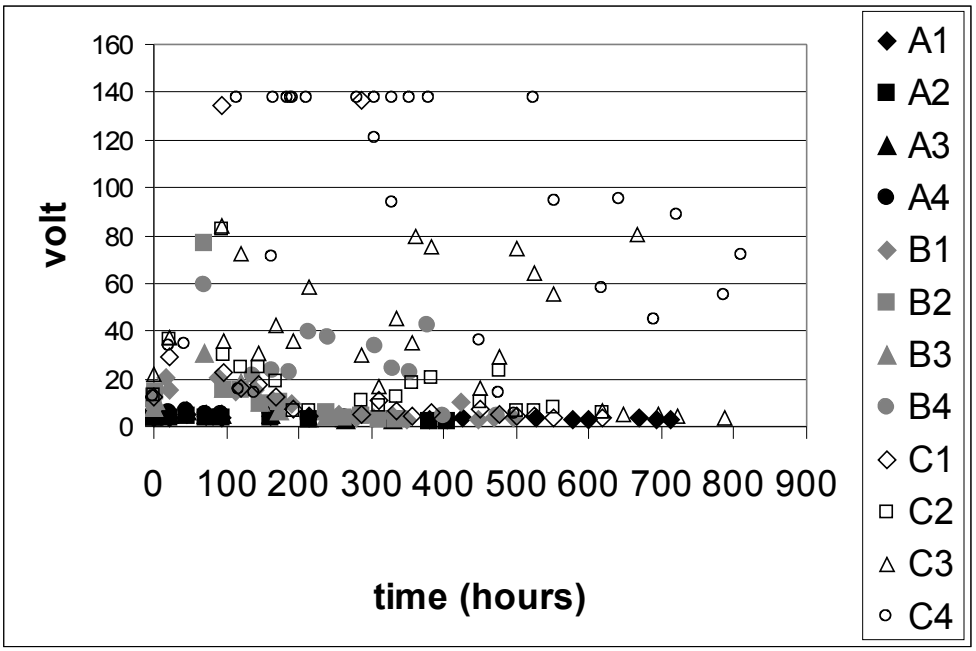

Figure 3.6: Voltage as a function of time in the 12 experiments

3.2.3 Pb-removal

In the 12 experiments, between 21% and 96% of the Pb was removed from chamber II into electrolytes or precipitated in membranes and at electrodes. Pb remaining in the liquid in chamber II constituted up to 43% of the total Pb after the end of the experiments. Final soil Pb-concentrations, ranging between 42 and 753

mg/kg, were obtained, showing how remediate of soil-fines was possible with the suspended EDR-setup.

An expected effect of acidification was that current-efficiency decreased with time due to production of H^+-ions competing with Pb^{2+} for transport. Therefore longer acidification-times per gram of soil were expected to result in an increase of current-efficiency for Pb-removal. Indeed this was seen in the experiments not affected by water splitting at the cation-exchange membrane (A1 through B3 and C1). In addition, when looking at the remediation rate (hours pr. mg Pb removed), the experiments with the longer acidification-time (A4 and B3) showed better remediation results than those with very fast acidification when looking at the remediation rate (hours pr. mg Pb removed). Therefore in the search of the most efficient remediation, it is not necessarily the fastest acidification which is preferential. Considering the total Pb removal, experiments affected by water splitting at the cation-exchange membrane (B4 and C2-C4) showed superior due to the longer remediation times. In addition the low concentration of ions in the liquid phase of chamber II of these experiments resulted in immediate removal of any ions released and higher removal of Pb from the soil solution.

3.2.4 Final distribution of Pb

Figure 3.7 shows the final distribution of Pb in the experimental cells. The majority of the Pb was transported towards the cathode as Pb^{2+}. Only in experiments where a large fraction of the Pb remained in solution in chamber II, Pb was found in small amounts in the anolyte, and Pb appears to have been transferred into the anolyte as co-ions. Both at the anode- and the cathode-sides a negligible fraction of the Pb

was found in the membranes. At the anode-side all Pb was found dissolved in the anolyte, while at the cathode-side the major Pb-pool was found in the catholyte in experiments A1, A2 and precipitated at the cathode as a porous substance in the remainder experiments.

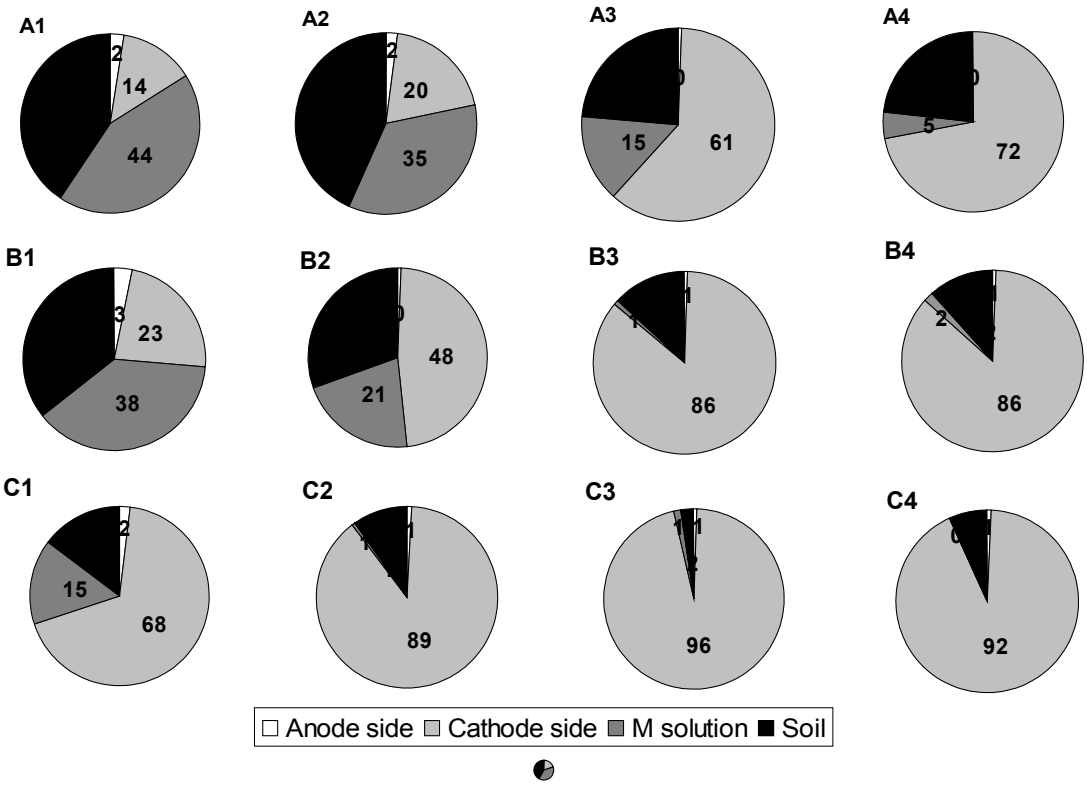

Figure 3.7: Final distribution of Pb in the 12 experiments (%).

3.2.5 Influence of time

At figure 3.8 the relation between final Pb concentration and remediation-time is visualized. Time pr. g. of soil is thought to be a crucial parameter in the case of up-scaling and commercial benefit, because it relates directly to residence-time and thereby size of equipment. It is evident that a relation exists; however, some

experiments show higher removal-rates than other. The experiments with the lowest removal-rates (points above the line in figure 3.8) are A1, A2 and C4. A1 and A2 due to the low current and C4 due to ion-deficiency caused by the combination of high L/S and high current. The experiments that show faster remediation than average (points below the line in figure 3.8) are B3, C1 and C2.

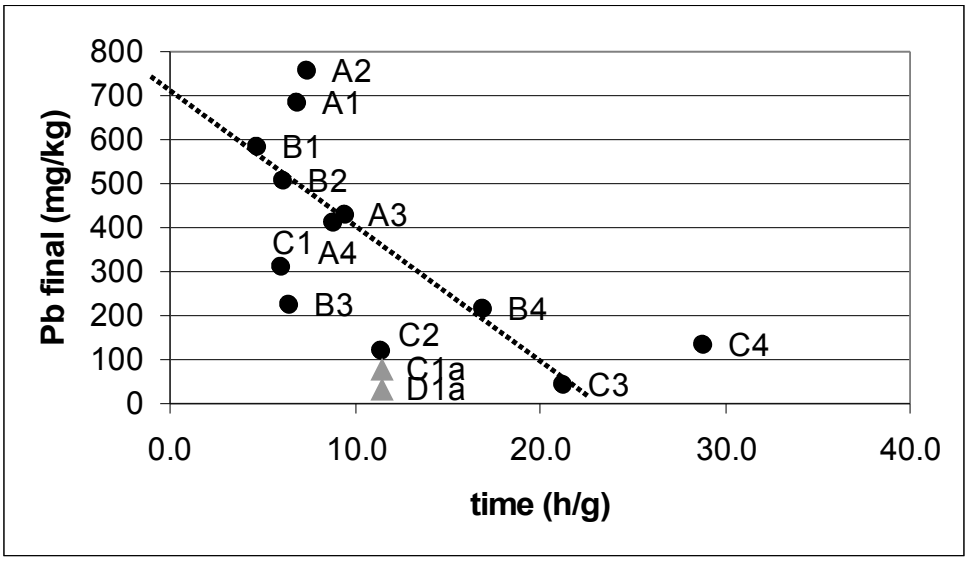

Figure 3.8: Relation between remediation-time (hours/g soil) and final Pb concentration (mg/kg). Initial Pb- concentration ≈ 1170 mg/kg.

Based on these findings, it was decided to repeat experiment C1 (experiment C1a) however, now running for 11.5 hours/gram soil for direct comparison with experiment C2. An additional experiment with further increased current density and high L/S (D1a) was also run for 11.5 hours/gram soil (see table II for details). Results of these two experiments are included in figure 3.8. The result of experiment D1a showed that remediation is possible to levels below the

governmental limit set by the Danish EPA for sensitive land use (40 mg/kg). Results of experiments C1 and C1a show how the removal rate decreases as the removal proceeds, because increasing amounts of H^+-ions compete for the transport. In consistence with this, current efficiency also decreases. In order to remediate efficiently it could therefore be beneficial to apply a number of reactors in series, where the initial reactor works at the highest possible removal rate, and the final reactor works at the target Pb-concentration. As conductivity increases with time the removal rate could be increased in secondary reactors by increasing current.

TABLE V

Results of experiments C1a and D1a

Exp. No.	Mass-balance [%]	Current efficiency [‰]	Pb removed from II [%]	Pb in liquid in II [%]	Hours pr. mg Pb removed	Final Pb soil conc. [mg/kg]
C1a	111	0.3	95	1.5	10.7	78
D1a	104	0.2	100	0	11.0	34

In table VI the optimal experiments at each L/S-ration are highlighted. It appears that the optimal current density increases linearly with decreased L/S in this region with the relation:

$$J_{opt} = 0.057(L/S) + 1 \quad (2)$$

TABLE VI

Dependency of optimal current density (bolded) on L/S.

L/S J [mA/cm2]	3.5	7.0	10.5	14.0
0.2	A1	A2	A3	**A4**
0.4	B1	B2	**B3**	B4
0.6	C1	**C2**	C3	C4
0.8	**D1**	-	-	-

4 CONCLUSIONS AND FUTURE RECOMMENDATIONS

With EDR in suspension, it is possible to remediate soil-fines completely, even from a soil with a high carbonate-content. During EDR of soil-fines in suspension, the lack of free anions results in water splitting at the anion-exchange membrane, resulting in acidification of the soil-slurry and mobilization of Pb. At high current densities and/or L/S, the lack of free cations results in water splitting at the cation-exchange membrane, resulting in production of hydroxide-ions and impeding the acidification of the soil. Water-splitting and remediation are highly dependent on L/S and current density. The optimal current density decreases linearly with increased L/S in the investigated region. The most efficient remediation is obtained when applying a current just below the limiting current for the cation-exchange membrane. Best results considering remediation rate were obtained at L/S 3.5 and current density of 0.8 mA/cm^2. Voltage can be used as a control-parameter for application of the ideal current.

In this experimental setup water splitting can be avoided by keeping a voltage-drop between the working electrodes below 20. The removal rate decreases as the removal proceeds. In order to remediate efficiently it could therefore be beneficial to apply a number of reactors in series, where the initial reactor works at the highest possible removal rate, and the final reactor works at the target Pb-concentration at an increased current density.